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PARTICLE SIZE ANALYSIS R. (U) ARIZONA STATE UNIV TEMPE
DEPT OF MECHANICAL AND AEROSPACE ENG.

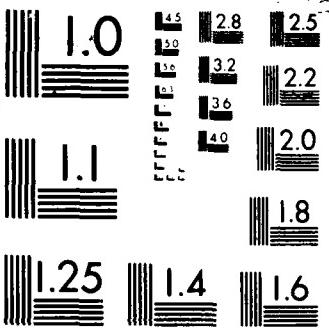
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19. Abstract

The fundamental scientific deficiencies impeding the integration of laser diffraction particle sizing techniques into intelligent sensors for next-generation propulsion systems have been identified. The research addressed three relevant areas: inverse scattering algorithms; multiple scattering; and the problems of laser beam deflections due to refractive index gradients in hostile propulsion environments. Direct integral transform techniques for the inverse problem have been formulated on a common basis and comprehensively evaluated. An error in the original derivation of the method of Petrov has been corrected and a new integral transform solution has been developed. Also, a scaling law for optimal configuration of laser diffraction detector geometries and size class characteristics has been developed which optimizes the stability and computational requirements of the inverse problem. Concerning multiple scattering, a new synthesis of successive order and discrete ordinates models has been developed for predictions of near-forward radiative transfer in optically thick media and the results have been verified with experimental data and an independent Monte Carlo simulation. Finally, a new concept involving programmable spatial filtering at the transform plane which allows annular detectors of variable geometries to be configured on-line by an intelligent instrument has been proposed and proof of principle experiments have begun. The concept will permit a detector to be automatically reconfigured around the deflected beam center and also will allow adaptive grid methods to be incorporated into the inversion algorithms.

RESEARCH ON CERTAIN ASPECTS OF LASER DIFFRACTION PARTICLE SIZE ANALYSIS RELEVANT TO AUTONOMOUS SELF-DIAGNOSING INSTRUMENTATION

Annual Report for 1 Oct 1985 - 1 Oct 1986
AFOSR Grant 84-0187

By

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Research Objectives

Particle and droplet size distributions, being parameters of fundamental importance, should be priority measurement objectives for intelligent sensors in next generation propulsion systems. Unfortunately there are a number of problematic scientific issues impeding the development of laser light scattering particle sizing instruments capable of on-line, autonomous, and self-diagnosing operation in hostile environments. The objective of this research effort is to contribute to the scientific knowledge base necessary to characterize and hopefully extend the applicability of laser diffraction instruments under these adverse conditions. Both the novel specific approaches discussed below and the emphasis on concepts relevant to intelligent instruments and sensors make this research project unique.

Our concept of a next-generation laser diffraction system is shown in Fig. 1 where the light scattered in the near-forward direction (i.e. diffracted) by particles along the line-of-sight of the probe laser beam is collected by a lens and directed to the back focal (Fourier transform) plane. The diffraction reference leg, variable focal length transform lens, beam position detector, programmable transmission filter, and a field detector to transduce the light passing through the transmission filter are novel concepts proposed here to extend the standard configuration utilizing a fixed focal length transform lens and a fixed geometry detector. We envision an intelligent instrument which performs self-calibration, self-diagnosis, and monitors the operating environment in order to automatically reconfigure both the "detector geometry" (i.e. the pattern in the transmission filter) and the inverse scattering algorithm to maintain operation near an optimal state. In the original proposal submitted to AFOSR in 1984 we identified three research areas in which the lack of scientific understanding would limit future advances. In the following paragraphs we discuss the research objectives associated with each of these target areas.

1. Inverse Scattering Algorithms

Particle sizing using laser (Fraunhofer) diffraction is an inverse scattering problem. The governing equation for single scattering by large particles is a Fredholm integral equation:

$$I(\theta)\theta^2 = I_{\text{inc}} \lambda^2 / 4\pi^2 \int_0^\infty J_1^2(\alpha\theta) \alpha^2 n(\alpha) d\alpha \quad (1)$$

where: $\alpha = \pi d/\lambda$ is the particle size parameter; $n(\alpha)$ is the unknown particle size distribution; and $I(\theta)$ the scattering intensity which can be measured at some number of scattering angles. Fredholm integral equations such as Eq. (1) are generally ill-posed in the sense that small variations in the measurements of $I(\theta)$ (e.g. due to noise) can cause large changes in the resulting $n(\alpha)$. The maximum information obtainable about $n(\alpha)$ is dependent on the noise levels in the measurements, the sampling or detection scheme, the stability or robustness of the inversion method, the actual size distribution, and the allowable level of uncertainty. Ideally, then, a laser diffraction system would have variable detector geometry, inversion scheme, and size resolution which could all be adjusted at run time depending on the conditions encountered.

The objective here is to identify and impact the underlying scientific issues which presently impede the needed quantum improvements in efficiency and robustness of inversion schemes for intelligent laser diffraction sensors. Dynamic adaptive control of the detector configuration and synergism of the inversion scheme with the detector geometry are focus areas.

2. Multiple Scattering

A major challenge for future research on *in-situ* optical techniques for particle sizing is to extend the applicability of these methods to optically thick media. The theoretical framework for particle size measurements under multiple scattering conditions is rather tenuous, even for the direct problem of calculating the radiation transfer properties of an arbitrary particulate medium. Even less developed are analytical methods for the inverse problem, *i.e.* obtaining particle size distribution information from measured optical scattering properties of media where multiple scattering is significant.

The objective of this part of the research is to understand the process of multiple scattering as it might occur in future applications of intelligent particle size distribution sensors and develop inversion schemes which can operate in such an environment. A minimum requirement is that multiple scattering be diagnosed by the instrument to ensure that erroneous data not be used in control algorithms, a situation which could result in a catastrophic failure. Our objective is to surpass that and develop efficient and robust algorithms which can actually extract useful particle size information from Fraunhofer diffraction measurements in multiple scattering environments.

3. Laser Beam Deflection/Steering

Another problem which has already limited the usefulness of laser diffraction particle sizing instruments in propulsion



systems research is that of beam deflection by refractive index gradients in the optical path. These gradients are due to spatially nonuniform temperature and/or species concentrations and at relatively large scales have the same effect as an array of randomly dispersed and time-varying lenses which cause the probe beam to move around at the detection plane. This has been a severe problem since information on the largest droplets in typical sprays is concentrated at small scattering angles which are of the same order as the beam deflection angles encountered.

The objective for this research topic is to investigate concepts which could enable a laser diffraction instrument to operate under conditions where beam deflection is significant. A minimal requirement is for on-line detection of beam steering so that an intelligent instrument would at least know that the data taken under these conditions is suspect. A more important objective is to develop strategies to permit a diffraction system to autonomously perform real-time correction for beam steering.

Status of the Research Effort

Progress has been made in each of these areas over the duration of this grant and we foresee no significant deviations from the original approach proposed for the research. The sections below describe the technical achievements made in these areas during the indicated 12 month time period.

1. Inverse Scattering Algorithms

The inverse scattering problem for laser diffraction particle sizing instruments has similarities with all systems requiring a deconvolution of experimental data. The inverse scattering problem (i.e. determination of $n(\alpha)$ in Eq. (1) from measured $I(\theta)$) can be solved by either (i) direct integral transform methods or (ii) indirect numerical quadrature approaches. In the past year we have continued a concentration on the generally overlooked integral transform techniques which, by virtue of the direct nature, have unique potential for very fast solutions as would be required by an on-line, intelligent sensor. We have completed a thorough study and synthesis of the integral transform formulations applicable to the inverse Fraunhofer diffraction problem and identified five unique methods. Three of these previously existed in the literature, one had errors in derivation which we corrected, and the fifth was newly developed as part of this work. A comprehensive parametric study of the important factors which control the inversion performance of the techniques has been completed and is detailed in the references.

Another important contribution this year was the development of optimal scaling laws for design of the detector geometry and size class characteristics of a Fraunhofer diffraction particle sizing system. Optimization here is in the sense that the computational requirements (storage and number of computations) for the inversion are minimized and the condition number of the

instrument function matrix is minimized (i.e. the stability of the numerical inversion or deconvolution is maximized). From inspection of the left hand side of Eq. (1) we propose that the detector signals S should respond as $S = I(\theta)\theta^2$. Numerical quadrature on Eq. (1) gives:

$$S = C \cdot A \quad (2)$$

where S , A , and C are matrices such that S_{jj} is the scattering signal on the j th detector, A_{ii} is the particle area in the i th size class proportional to $n(\alpha)\alpha^2$, and C_{jj} for the simplest quadrature scheme is given by:

$$C_{jj} = J_1^2(\alpha\theta) \quad (3)$$

Now the detector condition can be achieved using ring detectors where each annular element has a constant ratio of inner radius r_i to outer radius r_o such that each of the j th detectors has:

$$r_{i,j} / r_{o,j} = \theta_{i,j} / \theta_{o,j} = c \text{ (a constant)} \quad (4)$$

which also implies that the geometric mean angles of adjacent detectors have the same ratio. Now if the mean sizes of the size classes are also selected such that $\alpha_i / \alpha_{i+1} = c$ then:

$$C_{jj} = J_1^2(c^{i+j} \alpha_o \theta_o) \quad (5)$$

whereby all elements on any diagonal in the instrument function C matrix are equal, and there are only $2n+1$ unique elements in C (where n is the number of detectors and size classes). Clearly this cuts drastically the computation complexity as the storage requirements have decreased from $O(n^2)$ to $O(n)$. Further, if the α size classes are selected to maximize the signal S on a corresponding detector (e.g. $\alpha = 1.862/\theta$ to maximize J_1^2), then the C matrix has the largest elements on the diagonal and for small enough n will always be diagonally dominant.

2. Multiple Scattering

The specific problem of multiple scattering from particles large compared to the wavelength (i.e. Fraunhofer diffraction) has previously been modeled using successive orders and a combination of discrete ordinates and the adding method. We have developed a unique formulation which combines the method of successive orders with a discrete ordinates approach adopted specifically to the axisymmetric ring detector configuration used in most laser diffraction systems. Defining $S_n(\theta)$ as the probability that a photon scattered exactly n times will exit the medium within a finite scattering angle range represented by Θ we can write:

$$S(\theta) = \sum_{i=0}^{\infty} f_n S_n(\theta) \quad (6)$$

where S with no subscript represents the composite scattering signature which is the superposition of contributions from all scattering orders as would be measured by a laser diffraction instrument, and the occupancy variable f_n represents the probability that a photon undergoes exactly n forward scattering events. Now if the scattering event of interest is the n^{th} for a particular photon, then the scattering order signature $S_n(\theta_{\text{scat}})$ for photons forward scattered exactly n times can be found from:

$$S_n(\theta_{\text{scat}}) = \sum_{\text{all } \theta_{\text{inc}}} h(\theta_{\text{scat}}, \theta_{\text{inc}}) S_{n-1}(\theta_{\text{inc}}) \quad (7)$$

where h is the scattering redistribution function or the probability that a photon incident in direction θ_{inc} (for any azimuthal angle Φ_{inc}) will leave the next scattering event traveling in the direction θ_{scat} . The matrix h is a function of the size distribution and it is the efficient calculation and/or storage of h that is required for forward and inverse multiple scattering solutions.

Fig. 2 shows a comparison of our theoretical predictions with some experimental measurements of diffraction signatures from multiple scattering media, and the agreement is quite good. The effects of multiple scattering on apparent (i.e. as would be measured by a laser diffraction instrument) mean particle size is shown in Fig. 3. Here again the theoretical predictions from this study show good agreement with experiment.

These results indicate that a valid model for predicting multiple scattering signatures from media of arbitrary optical thickness has been developed. The formulation is optimized for inclusion into numerical schemes for inversion of multiple scattering measurements, and our research for the next year will concentrate on development and validation of inverse scattering formulations applicable to media of arbitrary optical depth.

3. Laser Beam Deflection/Steering

The probe laser beam from a Fraunhofer diffraction instrument will, in general, be continuously deflected as it traverses a medium of nonhomogeneous refractive index as would be encountered in a combustor. This causes the transmitted beam and the associated diffraction pattern which is centered around it to be directed to off-center points in the transform plane of Fig. 1. The very high intensity in the transmitted beam, which typically would be more than 100x greater than the intensity scattered into the small angle inner rings of the detector, severely hampers the measurements at small angles. Our concept for circumventing this problem is to use a programmable spatial filter at the transform plane. Annular transmission windows are configured about the instantaneous beam position as measured by the x-y position detector of Fig. 1, and light transmitted through the aperture is collected by the field detector. Our prototype system uses an array of Faraday effect transmitting

pixels and experiments to demonstrate the feasibility of this concept are underway.

Technical Publications, 10/1/85 - 10/1/86

1. Koo, J. H. and Hirleman, E. D. "Investigation of Integral Transform Techniques for Laser Diffraction Particle Size Analysis," Paper No. 77, Presented at the 1985 Technical Meeting, Eastern Section of the Combustion Institute, Philadelphia, PA, November, 1985.
2. Koo, J.H. and Hirleman, E.D., "Mathematical Formulation of Integral Transform Techniques for Laser Diffraction Particle Size Analysis," IR-86-11, Laser Diagnostics Laboratory, Mechanical And Aerospace Engineering Department, Arizona State University, Tempe, AZ, May, 1986.
3. Koo, J.H. and Hirleman, E.D., "Comparative Study of Laser Diffraction Droplet Size Analysis Using Integral Transform Techniques: Factors Affecting the Reconstruction of Droplet Size Distributions," Paper No. 86-18, presented at the 1986 Joint Spring Meeting of the Canadian and Western States Sections of the Combustion Institute, Banff, Alberta, CANADA, April, 1986. Under review for publication by the First International Congress on Particle Sizing, Rouen, France, May, 1987.
5. Hirleman, E.D., "Modeling of Multiple Scattering Effects in Fraunhofer Diffraction Particle Size Analysis," Paper No. 17, presented in the 1986 Joint Spring Meeting of the Canadian and Western States Sections of the Combustion Institute, Banff, Alberta, CANADA, April 28-30, 1986. Under review for publication by the First International Congress on Particle Sizing, Rouen, France, May, 1987.
6. Koo, J.H. and Hirleman, E.D., "An Improved Integral Transform Technique for Laser Diffraction Particle Sizing - Modified Petrov Method," To be submitted for publication in *Applied Optics*.
7. Koo, J.H., "Particle Size Analysis Using Integral Transform Techniques on Fraunhofer Diffraction Pattern," D.Sc. Dissertation, George Washington University, Mechanical Engineering Department, Washington, DC, in preparation.
8. Hirleman, E. D. "Optimal Scaling for Fraunhofer Diffraction Particle Sizing Instruments", under review for publication by the First International Congress on Particle Sizing, Rouen, France, May, 1987.
9. Hirleman, E. D. "Programmable Transform Plane Filter for Dynamically Configurable Fraunhofer Diffraction Detector", in preparation, to be submitted for publication to *Applied Optics*.

Professional Personnel

Prof. E. Dan Hirleman - Associate Professor of Mechanical and Aerospace Engineering

Joseph H. Koo - Research Assistant and Ph.D. student. Mr. Koo's dissertation based on this research is written and his Ph.D. should be awarded in early 1987.

Interactions

1. June, 1986 visit by Prof. E. D. Hirleman to Kirtland Air Force Base, Albuquerque to visit Dr. Tim Ross and Mr. Glenn James based on preliminary contacts at the AFOSR Contractor's meeting at Stanford in June. Discussions centered on particle diagnostics requirements for research underway at Kirtland.

New Discoveries

1. Optimal Scaling for Fraunhofer Diffraction Particle Sizing Instruments

We have derived scaling laws for the optimal design of a Fraunhofer diffraction particle sizing system. The scaling laws apply to the detector geometry and the size class characteristics and are optimal in the sense that the computational requirements (storage and number of computations) for the inversion are minimized and the condition number of the instrument function matrix is minimized (i.e. the stability of the numerical inversion or deconvolution is maximized). Further, the same scaling laws will have a significant impact on compressing the data storage and computations required for the inverse multiple scattering solutions we are developing. Further details on this innovation can be found in ASU Patent Disclosure No. 212 and the referenced paper by Hirleman on this topic.

2. Programmable transform plane spatial filter for dynamic annular detector configuration.

Another invention derived from this research is the concept of a programmable transform plane spatial filter for dynamic configuration of annular detectors as shown in Fig. 1. This concept will allow incorporation of significant levels of intelligence into an instrument including on-line control of the ring detector center (for beam deflection) and the detector geometry with capabilities for adaptive grid and variable resolution configurations. Further details on this innovation can be found in ASU Patent Disclosure No. 213 and the referenced paper by Hirleman on this topic.

3. Successive order model for multiple scattering.

We have developed a new model for the prediction of the forward scattering characteristics of optically thick media where

the particles are large compared to the wavelength. The formulation is based on a new synthesis of successive order and discrete ordinates methods and is optimized for inclusion into inverse multiple scattering problem. The formulation produces the relatively simple relation:

$$S = \exp(-b) \exp(b/2 \cdot H) S_0$$

where S_0 is the vector representing the angular distribution of the input optical energy to the medium (all at $\theta = 0$ for collimated beam input) and S and H are the scattering vector and the redistribution matrix as defined in the report above. Note that the direct calculation of the scattering signature S is reduced to knowledge of the input laser angular spectrum S_0 and exponentiation of the redistribution matrix H . Further, this equation can be rearranged to provide for an experimental determination of H which can then be used to determine $S_1(\theta_{scat})$ (scattering signature of single scattered photons) from which the size distribution can be determined using conventional single scattering inversion methods. Further details on this innovation can be found in ASU Patent Disclosure No. 214 and the referenced paper by Hirleman on this topic.

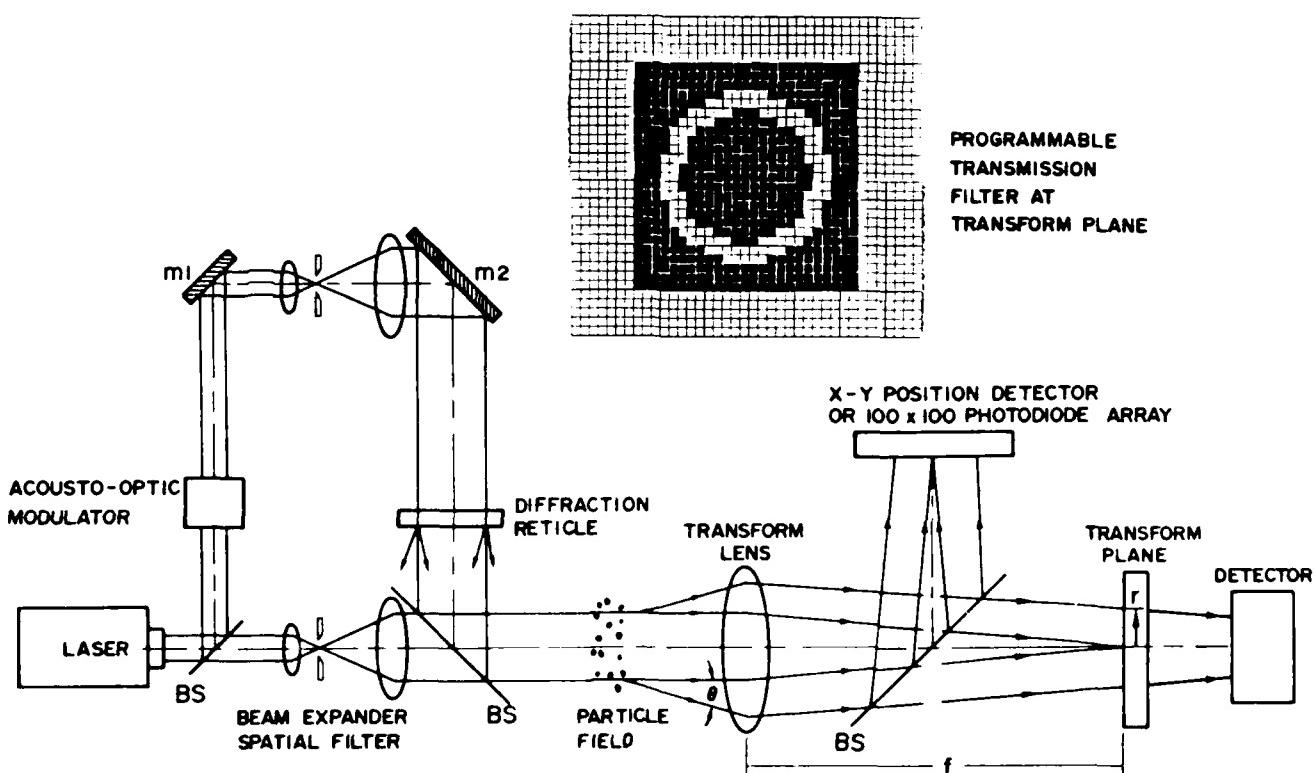


Figure 1. Schematic of a next-generation laser diffraction particle sizing system for use as an intelligent sensor.

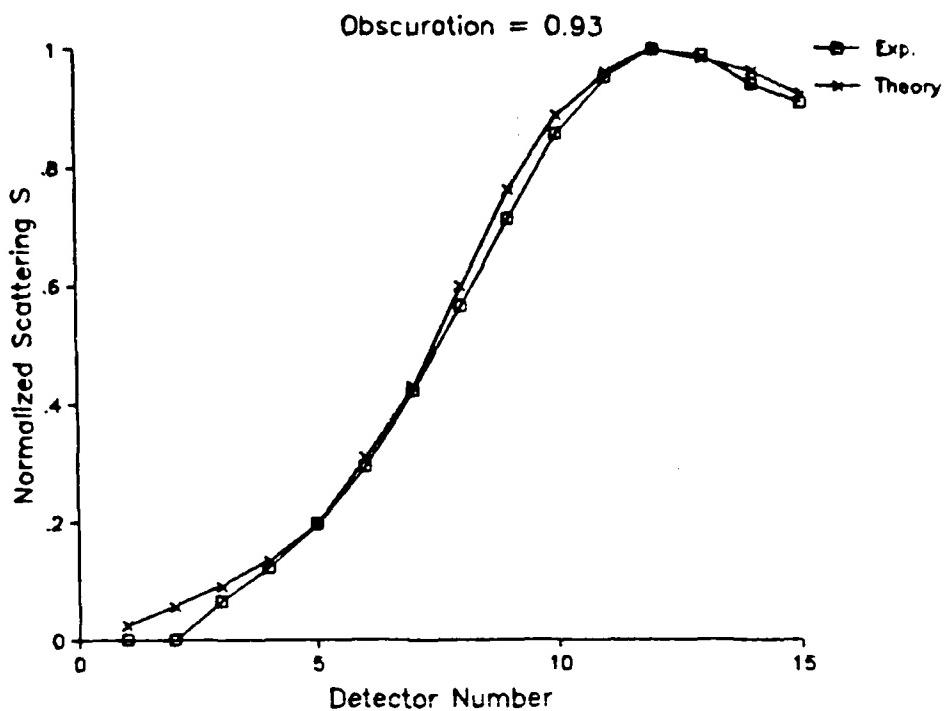


Figure 2. Scattering signature vs. detector number (RSI/Malvern) for an optically thick spray. Predictions from the multiple scattering model developed here are compared with experimental results obtained by Dodge (*Optical Engineering*, V. 23, p. 626, 1984).

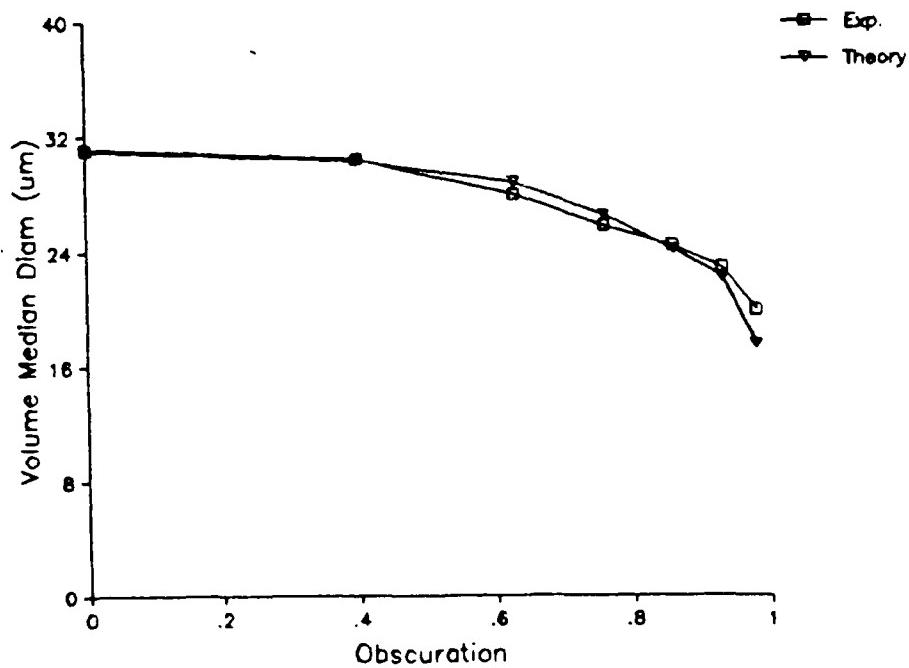


Figure 3. Volume median diameter $D_{v,0.5}$ as measured by a laser diffraction system for various optical thicknesses of the spray of Fig. 2. compared with multiple scattering predictions.

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